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LOW ENERGY INELASTIC ATOMIC AND MOLECULAR COLLISIONS.(U)

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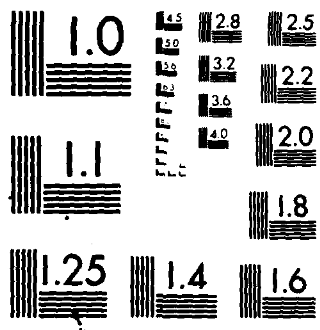

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20. ABSTRACT CONTINUED

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Reliable grazing-incidence soft x-ray spectra were taken for the first time showing multiplet and charge-state structure in the 60-100Å range from collisions of intense 80-150 keV P^{+} and S^{+} ion beams with an Ar gas target. The spectra are dominated by $L_{2,3}$-M and $(L_{2,3})^2$-LM satellite lines of the projectile ions corresponding to very high degrees of outer-shell ionization (fluorine-like to aluminum-like states). Auger cascades from the initial $(2p^{-2})$ vacancy-states produced in the primary collision appear to be important.

Final Report: Low Energy Inelastic Atomic and Molecular Collisions - Grant DAAG-29-79-C-0125

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15 April 1981

Abstract

Ar^{++} + Ar collisions are studied with particular emphasis on the single electron capture channels. The dominant process is found to result in $Ar^{+}(^{2}P) + Ar^{+}(3s^{2}3p^{4}n\ell)$. A weaker process, attributed to the presence of a highly excited Ar^{++} state is also seen in the angular range investigated.

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at ~72Å and 91Å are discussed. Wavelengths of several new soft x-ray lines, not seen in electron excitation, from $2p^4 3s^m 3p^n$ and $2p^5 3s^m 3p^n$ configurations have been measured for the first time. Additional work on collision-induced 500-1100Å spectra of Ar and Ar^+ from He^+ and Ne^+ projectiles is discussed, including apparent population inversion on the 924Å doublet of Ar^+.

I. Ion-Atom and Ion Molecule Interactions

A. $Ar^{++} + Ar$ Collisions

The $Ar^{++} + Ar \rightarrow Ar^{++} + Ar$, $Ar + Ar^{++}$, and $Ar^{+} + Ar^{+}$ collisions are investigated in the low keV energy range for angles out to one degree. The direct scattering is found to be primarily elastic in the $\tau<3$ keV deg range studied with almost no evidence of Ar excitation. The results for two electron capture suggest resonant charge exchange as the dominant process for this channel. The $Ar^{++} + Ar \rightarrow Ar^{+} + Ar^{+}$ collision shows two processes (A and B). The dominant process, B, is attributed to $Ar^{++}(^3P) + Ar \rightarrow Ar^{+} + Ar^{+}(3s^2 3p^4 n\ell)$.

The weaker process, A, is tentatively assigned to states such as $Ar^{++}(^1P^o)$ and $Ar^{++}(^5D^o)$ that lie about 18 eV above the ground state and can populate highly excited Ar^{+} states. The results are discussed in more detail in Appendix 1.

B. Ion-Molecule Collisions

The $D^{+} + H_2$ collision is studied at low keV energies in the $E_o\theta^2<3$ keV deg^2 range. The location of the peak for the quasi-elastic scattering shows that rotational excitation of the H_2 electronic ground state is the important contributing process. Although vibrational excitation is allowed for $E\theta^2>1.5$ keV deg^2 no direct experimental evidence for the opening of vibrational channels is found. Electronic excitation corresponding to $D^{+} + H_2^{*}$:$Q\simeq 13$ eV is seen over a limited $\tau=E\theta$ range and the results suggest excitation of this process via a curve crossing mechanism. Charge exchange is found to be important in this collision system. The results are discussed in a forthcoming Physical Review paper.

A model was proposed to explain our earlier charge exchange results in $He^{+} + H_2$. The model is based on a crossing between the incident channel and an exchange channel leading to $H_2^{+*} + He^{*}$. This is discussed in Appendix 2.

II. Soft X-ray and Vacuum-Ultraviolet Emission Spectroscopy from Collisions.

A. X-rays

At the beginning of this grant period, our 150 keV Cockcroft-Walton ion accelerator was replaced with a custom research version of the Varian-Extrion Model 200-DF4 ion implantation accelerator, capable of high current densities of mass-analyzed ions in the 10-200 keV range approaching 1 mA/cm^2 DC. An improved target chamber, suitable for either gas or solid targets was installed on the beam line of this accelerator at the entrance slit of our 1m grazing-incidence XUV-soft x-ray monochromator. Improved differential pumping and a new gas cell design (permitting a higher range of operating pressures), together with the higher current capability of the accelerator, allowed us for the first time to obtain reliable grazing-incidence spectral measurements of the multiplet structure in the soft x-ray emission from keV-energy light ion-atom collisions with a gas target. The details of these successful measurements were presented at conferences during 1980 and 1981 and are given in the appended abstracts (App. 3 and 4).

Another paper will be published shortly in the IEEE Transactions on Nuclear Science (1981).

The intent of this work, described in our proposal, was to obtain sufficient resolution of phosphorus, sulfur and argon L-x rays under single collision conditions so that specific transitions could be identified, including multiplet structure and charge-state information. Coupled with theoretical calculations of the L-shell fluorescence yields (the ratio of decay by photon emission to all decay modes), the data can then be interpreted in terms of the specific initial vacancy states produced in the primary ion-atom collision. Successful single-collision spectra were obtained for P^{+}+ Ar at 100 keV and S^{+}+ Ar at several bombarding energies in the 80-150 keV range.

Data of the kind proposed on pages 34 and 35 of our proposal were obtained as follows: 1) projectile-ion spectra from phosphorus and sulfur excited in collisions with Ar gas were obtained and are now being analyzed to see if the Russell-Saunders term intensities for each observed configuration are given by the statistical-weight ratios or whether there is evidence of preferential excitation of specific angular momentum states. The gas target used in our apparatus contains a thin soft x-ray transmitting polypropylene window in addition to a window on the detector. The transmission of these windows must be accurately known in order to determine the desired intensity ratios. The data analysis will be completed as soon as this source of systematic error is understood. General indications at present do not favor large deviations from statistical ratios within most of the multiplets.

2) For asymmetric collisions, we have determined the relative x-ray intensities (subject to final corrections for window transmission) for L-shell x-ray lines corresponding to single and double L-shell vacancies produced in the collision. The molecular-orbital model in its simplest form predicts that the primary collision should produce primarily double vacancies ($4f\sigma$ orbital ionization) yet the spectra show strong lines involving only single L vacancies in the projectile 2p orbital. It is not yet clear whether this is due to fluorescence-yield differences between the L and L^2 x-ray transitions or to a breakdown of this simple model of the collision process. We plan to pursue this point further in consultation with Professor B. Crasemann's group at the University of Oregon. Because of much lower fluorescence yields for Ar projectile x-rays (compared with P and S), we have not yet been able to obtain high quality spectra for symmetric $Ar^+ + Ar$ collisions. The effort to do so will continue.

3) In both the $P^+ + Ar$ case and the $S^+ + Ar$ case, we find the soft x-ray spectrum in the 60-100 Å range to be dominated by $L_{2,3}$-M and $(L_{2,3})^2$-LM satellite lines corresponding to charge states implying high degrees of outer-shell ionization. In particular, the neon-like $2p^5 3s-2p^6$ unity fluorescence yield doublet is prominent in both cases, representing of the order of 1% of the total L-shell ionization cross section. Excitation of these transitions requires stripping 4 or 5 electrons from the outer (M) shell of the projectile, highly unlikely as a direct

process at 100 keV. The most probable excitation mechanism would appear to be an Auger cascade following double 4fσ L-shell ionization, which produces an increase in the number of M shell vacancies by two along with filling one of the L-shell holes. Furthermore, one of the members of this doublet is slightly metastable. This suggests the possibility that one might be able to use this transition to produce laser action in the 72 Å range in S^{6+} and in the corresponding 91 Å line of P^{5+}. That is, a population inversion may be feasible on these transitions but the small cross section would imply a low gain unless very high current density S^+ or P^+ beams are used. Recent developments in the production of short, high-current pulsed beams of light ions might make these collisions plausible candidates for ion-beam pumped lasers at some time in the future. We do not have the resources to pursue this line of research more intensively at the present time.

To summarize this aspect of our work, we have been able to resolve considerable multiplet and charge-state structure in the soft x-ray spectrum from collisions of P^+ and S^+ mass-analyzed beams with an Ar gas target in the 100 keV range where the soft x-ray cross sections and fluorescence yields are rising rapidly. Previous spectral measurements (by Fortner's group at Livermore, see Phys. Rev. A10, 2218 (1974)) on these collision systems using a curved crystal Bragg spectrometer instead of a high-throughput grazing incidence monochromator, were unable to resolve much of this structure. Nevertheless, their earlier work and absolute cross section measurements have been very valuable aids in analyzing our present data. Nearly all of the transitions observed have been identified as due to fluorine-like-aluminum-like configurations of the projectile ions. Excitation of the higher-Z target ion L x-rays is extremely weak, if present at all, consistent with the inelastic energy-loss measurements made by Fastrup and co-workers and also with the Livermore data.

The kinds of electron configurations seen in these multi-charged ion projectile spectra are very different from those seen with electron-beam excited x-ray spectra (for comparison see J. Nordgren _et al_, Physica Scripta _19_, 5 (1979)). A number of previously unidentified transitions involving either one or two 2p vacancies in the projectile ions were measured and tentatively identified. In sulfur, for example, there are several transitions of the type $2p^4 3s^m 3p^n (m+n>,2)$ in the 65-70 Å range and also new lines ascribed to $2p^5 3s^m 3p^n (m+n=2,3)$ in the 74-80 Å range. These transitions are only partially resolved at the 0.45 Å resolution used, but we believe the apparatus to be capable of at least a factor 2 resolution improvement with only minor modification. Theoretical calculations are not yet available for the fluorescence yields of all of these transitions, but we believe most of them are <1%, implying strong excitation of these new lines in the primary collisions.

The absence of transitions due to 2s vacancies in the projectile and L-shell x-rays of the higher Z partner implies that the dominant mechanism for excitation is probably $4f\sigma$ molecular-orbital ionization. Direct Coulomb ionization of these spectra is not expected to be important in the 100 keV range. The dominant excitations in both the $P^+ + Ar$ and $S^+ + Ar$ cases appear to be quite analogous, as expected from the MO model.

B. Vacuum Ultraviolet Spectra from 8-30 keV Collisions

Data obtained during the previous grant from AROD have been analyzed with the help of Dr. Kennedy J. Reed of Lawrence Livermore Laboratory. A paper on ion-atom rearrangement collisions involving excitation of the 3p-3s doublet of Ar^+ at 924 Å in collisions of He^+ and Ne^+ with Ar gas is nearly completed. Comparison of the data with several types of model calculations indicated that the excitation of this line occurs predominantly via charge-exchange or "vacancy-sharing" leaving the target ion neutralized, in the energy range studied. The model calculation that gave the best agreement with the data was a Demkov exponential-model calculation with translation-factors included (this brings down the calculated cross section at high energies). A Nikitin-model calculation, based on Dr. Reed's calculated quasimolecular potential curves for the $HeAr^+$ system and omitting the translation factors, gave similar results at low keV energies but gave too large a cross section above 20 keV. It appears likely that the cross section for the 924 Å Ar^+ doublet is dominated by ionization in the 100 keV range and above.

The large peak (approximately 10^{-16} cm^2) in the emission cross section near 9 keV, along with the much smaller Demkov-model cross section for charge exchange into the ground state of Ar^+, imply that a large population inversion ratio for $Ar^+(3s^{-1})$ production should be produced in the primary collision. Estimates of the gain to be expected in stimulated emission on this doublet were only moderately encouraging. For lasing on these transitions using ion beam excitation, high-current pulsed He^+ (or perhaps Ne^+) beams in the kiloampere/cm^2 range appears to be a minimum requirement. Nonetheless, this particular type of charge-transfer process may be worthy of further investigation.

The dominance of quasimolecular effects in both He^+ + Ar and Ne^+ + Ar collisions up to 30 keV is strongly suggested by the dramatic differences between the spectra from these collisions and from the "inverse" collisions of Ar^+ with He, Ne gas. The 924 Å doublet is almost totally absent in the inverse collisions. In the He^+ + Ar collision, there are other intense VUV lines as well: the dominant ones observed in the 500-1100 Å range were emission from the 2p excitation of neutral He (584 Å resonance line), 4s and 3d excitations in Ar^+, and weaker excitation of the 1048-1067 Å resonance lines of neutral Ar.

Although this particular investation of ion-induced VUV emission spectra is now terminated, the new beam line we are using for soft x-ray grazing incidence collision spectroscopy is also usable in studying ion-induced VUV emission with good resolution. All that is required is a simple change of grating and photon-counting detector. The same data-acquisition system and ion-implantation accelerator (purchased with industrial and internal University support) can be used in the VUV region. We plan to carry on additional studies in both the soft x-ray and VUV region as our resources permit. We also intend to extend these spectroscopic studies in the direction of solid metallic surfaces that have been subjected to ion implantation, as outlined in our original three-year proposal.

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Appendix I

Charge-exchange in small-angle $Ar^{++} + Ar$ collisions*

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Abstract

Charge-exchange in $Ar^{++} + Ar$ is studied at low keV energies and scattering angles out to one degree. The dominant one-electron process results in $Ar^{+}(^{2}P) + Ar^{+}(3s^{2}3p^{4}n\ell)$. A weaker process is attributed to the presence of highly excited Ar^{++} states. The results can be explained using an $Ar^{++} + Ar^{*}$ intermediate state that crosses the incident and final channels.

Introduction

There is currently substantial interest in understanding charge exchange processes involving multiply charged ions. This is in part due to the fact that such processes occur in highly ionized plasmas, and the corresponding cross-sections are important in determining the charge state distributions. In addition the exchange processes result in energy losses in the system by radiation from products left in excited states. Charge exchange in $Ar^{++} + Ar$ is particularly interesting and has been discussed in several recent papers [1-5] covering a wide energy range. In this system there is some difficulty in understanding the Ar^{+} states thought to be involved in single electron capture processes. Our laboratory is currently studying low keV energy-small angle $Ar^{++} + Ar$ collisions, and our results and interpretations are not in agreement with those of other recent studies. In particular in $Ar^{++} + Ar \rightarrow Ar^{+} + Ar^{+}$ we do not find that the dominant small angle processes result in $Ar^{+}(^{2}P) + Ar^{+}(^{2}P)$ and $Ar^{+}(^{2}P) + Ar^{+}(3s3p^{5})$.[1,2] The latter channel is difficult to justify and is not understood.[1] In this paper we show that the dominant one electron charge-exchange process is $Ar^{++} + Ar \rightarrow Ar^{+}(^{2}P) + Ar^{+}(3s^{2}3p^{4}n\ell)$ at small $\tau = E\theta$ (reduced scattering angle = beam energy x scattering angle). A model is presented[6] to explain excitation of these channels in the τ region studied.

Experimental Technioue

The basic apparatus has been described in an earlier paper[7] on collisional ionization in Ar + Ar, and the small modifications will be discussed in a future paper. In $Ar^{++} + Ar \rightarrow Ar^{+} + Ar^{+}$ the states are identified from ΔE, the measured energy loss or gain of the scattered Ar^{+}. Since there is no Ar^{+} reference peak from which ΔE can be determined, it is necessary to measure the Ar^{++} and Ar^{+} energies and use the known electrostatic energy analyzer constant to find ΔE. An additional problem is caused by contact potentials in the scattering cell and on the analyzer. Because of the change in charge state these potentials can yield ΔE values which may be in error by about 1 eV. In the present study both of the above problems are resolved by using $Ar^{++} + He \rightarrow Ar^{+} + He^{+}$ collisions (where the possible final states are far apart in ΔE) as a reference. Figure 1 shows composite spectra of $Ar^{++} + Ar \rightarrow Ar^{+} + Ar^{+}$ at $E = 2.8$ keV - $\theta = 0.15$ deg and $A^{++} + He \rightarrow Ar^{+} + He^{+}$ at $E = 2.8$ keV - $\theta = 0$ deg. The reference peak is assigned to $Ar^{++}(^{3}P) + He \rightarrow Ar^{+}(^{2}P) + He^{+}(^{2}S)$ with $\Delta E = +3$ eV (agreeing to within 1 eV with the value computed using the known analyzer constant). The reference peak is obtained at 0 deg to avoid the corrections for the kinematically required energy loss at non-zero scattering angles.[8]

$Ar^{++} + Ar$ Collisions

The $Ar^{++} + Ar \rightarrow Ar^{++} + Ar$, $Ar + Ar^{++}$, and $Ar^{+} + Ar^{+}$ collisions are investigated in the low keV energy range for angles out to one degree. The direct scattering is found to be primarily elastic, in the $\tau<3$ keV deg range studied, and shows almost no evidence of Ar excitation. This is in sharp contrast to the $Ar^{+} + Ar$ case where excitation of Ar(4s,4p) is found[9] in the same τ range. The angular dependence for the two electron capture process at E=1.7 keV is shown in Fig. 2. The oscillations strongly suggest resonant charge exchange as expected. The $Ar^{+} + Ar^{+}$ spectrum in Fig. 1 is of most interest. Using $Ar^{++}(^3P) + He \rightarrow Ar^{+} + He^{+}$, $\Delta E = Q = +3$ eV, as a reference (peak R), peaks A and B correspond to Q values (at the maxima) of about +5.6 and -6.7 eV respectively. Peak B at this angle suggests $Ar^{++}(^3P) + Ar \rightarrow Ar^{+} + Ar^{+}(3s^2 3p^4 3d)$. This peak shows Q values that depend somewhat on E and θ, but they are consistent with the assignment of a final $Ar^{+}(3s^2 3p^4 n\ell)$ state. The incident Ar^{++} beam is primarily in its ^3P ground state but contains ^1D, ^1S and other long-lived highly excited state components.[1,10] Peak A is tentatively assigned to states such as $Ar^{++}(^1P^o)$ and $Ar^{++}(^5D^o)$ that lie about 18 eV[11] above the ground state and can populate highly excited Ar^{+} states with Q values in the range found. Although the highly excited states are expected to constitute only a very small part of the Ar^{++} beam, large cross sections associated with these states can contribute significantly to "total cross-section" measurements and to emission spectra from $Ar^{++} + Ar$ collisions. Figure 1 suggests other processes, but they are found to be only weakly excited in the τ range studied.

Excitation of $Ar^{+} + Ar^{+}(3s^2 3p^4 n\ell)$ can be explained by a recently proposed model[6] using intermediate states, associated with $Ar^{++} + Ar^{*}$, that cross the incident and outgoing channels. Figure 3 shows several potential energy curves for the $Ar^{++} + Ar$ and $Ar^{+} + Ar^{+}$ systems. The curves for $Ar^{++} + Ar$ are plotted using a potential energy $V(r) = -2\alpha e^2/r^4$ where α is the polarizability of Ar ($1.64 \times 10^{-24} cm^3$),[12] e is the electronic charge, and r is the internuclear separation. The curve for $Ar^{++}(^3P) + Ar^{*}$ is plotted for $Ar^{*}(4s)$ with $\alpha = 48 \times 10^{-24} cm^3$ [13] and is displaced at infinite separation by 11.5 eV[11] above $Ar^{++}(^3P) + Ar$. Curves corresponding to several $Ar^{+} + Ar^{+}$ states are plotted using a Coulomb potential. Although only simple potentials are assumed to illustrate the model, they should be valid for $r > 3 Å$. Excitation of $Ar^{+} + Ar^{+}(3s^2 3p^4 n\ell)$ involves a two step process: (1) an initial crossing between the incident $Ar^{++}(^3P) + Ar$ and a state such as $Ar^{++}(^3P) + Ar^{*}$, (2) a transition (occurring near $r = 4.5 Å$) from this intermediate state to $Ar^{+} + Ar^{+}(3s^2 3p^4 n\ell)$. The $Ar^{++} + Ar^{*}$ is depopulated by crossings with the $Ar^{+} + Ar^{+}(3s^2 3p^4 n\ell)$ states, and the direct inelastic scattering should be weak (as found). Although the intermediate state also crosses $Ar^{+} + Ar^{+}(3s3p^5)$, at $r < 4 Å$, this channel should only be weakly excited since it involves a two electron transition at the crossing.

The Q values found for peak A do not allow its excitation by the ^3P, ^1D, or ^1S states in the Ar^{++} beam. To keep Figure 3 simple, the curves for peak A are not plotted, but the process can be understood by referring to the figure. The state (or states) responsible for the peak is assumed to lie about 18 eV above $Ar^{++}(^3P)$ + Ar at infinite separation. The curve is assumed to be parallel to $Ar^{++}(^3P)$ + Ar (it would be more attractive if a term dependent on the polarizabilities of the ground and excited states of Ar^{++} were included in V(r)) and crosses highly excited final states of Ar^{+}+ Ar^{+*} which are parallel to the Ar^{+}+ $Ar^{+}(3s^2 3p^4 n\ell)$ curve shown. These crossings occur at large r, and only one crossing is required for the charge exchange. As r decreases, the crossings allow population of lower-lying final states. This is in agreement with the experimental results which show an increase in Q value with increasing scattering angle.

Conclusion

The data suggest that the dominant one electron capture process, in Ar^{++}+ Ar collisions for τ<3 keV.deg., is of the type $Ar^{+}(^2P)$ + $Ar^{+}(3s^2 3p^4 n\ell)$. A weaker peak (except for angles very close to zero) is attributed to the presence of a highly excited Ar^{++} state in the incident beam. To date only exothermic processes could be understood in terms of a simple model where crossings occur between the attractive incident state and the final repulsive Coulomb states. Intermediate states of the type proposed can account for the observed endothermic channels at small τ value. The agreement between conclusions based on the proposed model and the data suggests that long range interactions should be considered in theoretical treatments of the problem.

Acknowledgement

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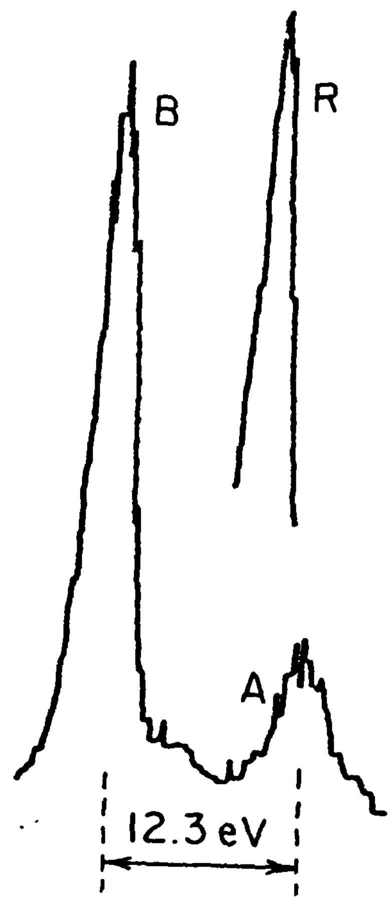
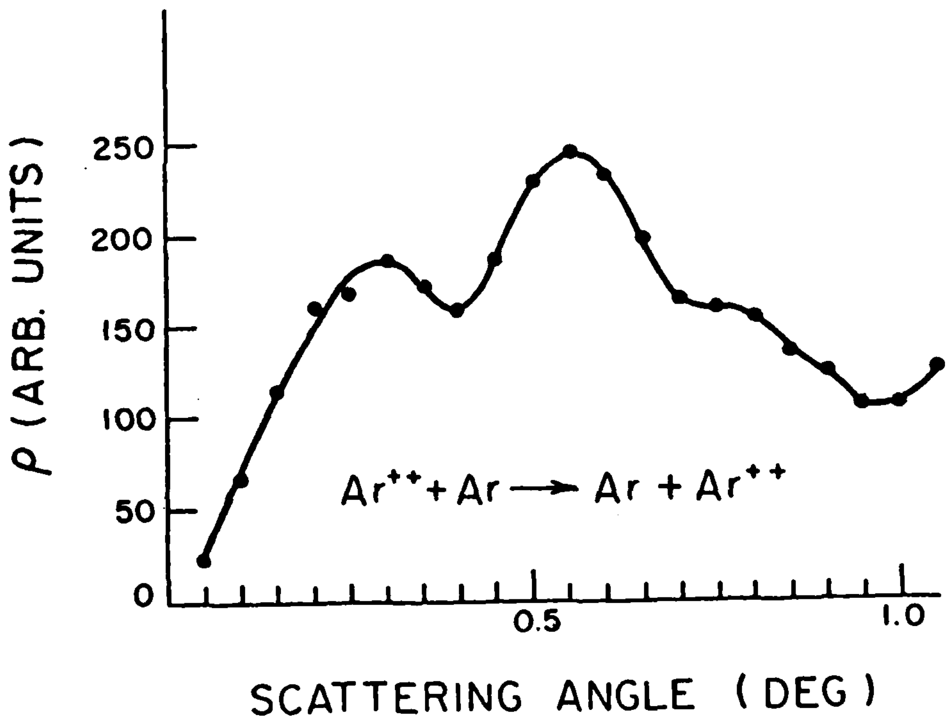
Figure Captions

Fig. 1. Composite spectra showing 2.8 keV $Ar^{++} + Ar \rightarrow Ar^{+} + Ar^{+}$ at 0.15 deg and $Ar^{++} + He \rightarrow Ar^{+} + He^{+}$ at 0 deg. Peak B is attributed to $Ar^{+}(^{2}P) + Ar^{+}(3s^{2}3p^{4}3d)$. At all angles this peak is consistent with final $Ar^{+}(^{2}P) + Ar^{+}(3s^{2}3p^{4}n\ell)$. Peak A, resulting from the presence of highly excited long lived states in the Ar^{++} beam gets weaker and moves toward larger Q values with increasing angle. Other weak processes are suggested by the spectrum. Peak R serves as a reference and corresponds to 2.8 keV-0 deg. $Ar^{++}(^{3}P) + He \rightarrow Ar^{+}(^{2}P) + He^{+}(^{2}S)$:Q=3 eV.

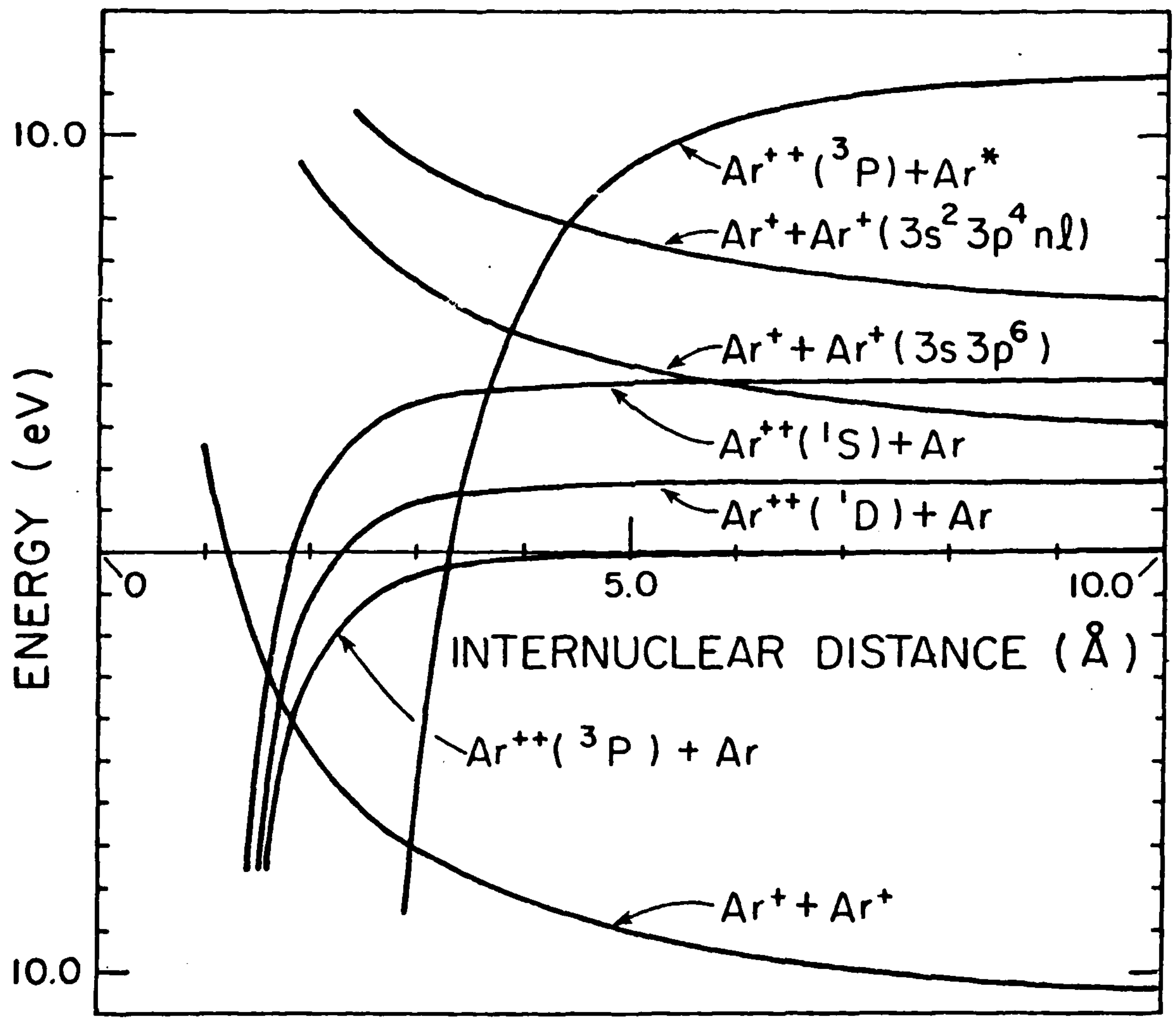
Fig. 2. $Ar^{++} + Ar \rightarrow Ar + Ar^{++}$ at 1.7 keV. The plot shows ρ (scattered beam intensity x θ^{2}) as a function of scattering angle and suggests a resonant charge exchange process.

Fig. 3. Potential energy curves for selected states in $Ar^{++} + Ar$ and $Ar^{+} + Ar^{+}$ to illustrate the proposed model. The spacing between states at infinite separation is obtained from Ref. 11. $Ar^{++}(^{3}P) + Ar^{*}$ corresponds to $Ar^{*}(4s)$. The $Ar^{+} + Ar^{+}(3s^{2}3p^{4}n\ell)$ is plotted for the lowest level which lies 16.4 eV above the $Ar^{+} + Ar^{+}$ ground state. All other states corresponding to $n\ell$ configurations lie above it. States to which peak A is attributed, and intermediate states such as $Ar^{++}(^{1}S)+Ar^{*}$ which may be important in the collision are not shown to keep the figure simple. Capture into $Ar^{+} + Ar^{+}$ from $Ar^{++}(^{3}P) + Ar$ (which is a weak process in the τ range studied) would result in a Q = +11.9 eV.


B
R
A
12.3 eV



250
200
150
100
50
0
0.5
1.0
ρ (ARB. UNITS)
Ar⁺⁺ + Ar → Ar + Ar⁺⁺
SCATTERING ANGLE (DEG)



10.0
ENERGY (eV)
10.0
0
5.0
10.0
INTERNUCLEAR DISTANCE (Å)
$Ar^{++}(^3P)+Ar^*$
$Ar^{+}+Ar^{+}(3s^2 3p^4 n\ell)$
$Ar^{+}+Ar^{+}(3s\,3p^6)$
$Ar^{++}(^1S)+Ar$
$Ar^{++}(^1D)+Ar$
$Ar^{++}(^3P)+Ar$
$Ar^{+}+Ar^{+}$

Appendix II

The Importance of Long Range Forces in Ion-Molecule Charge-Exchange Collisions: $He^+ + H_2^+$

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Abstract

The importance of long-range forces in $He^+ + H_2$ charge-exchange collisions is shown. A model is proposed where curve crossings due to the large differences in polarizabilities between the ground and excited atomic states result in exchange collisions.

In recent years a great deal of experimental and theoretical effort has been devoted to studies of charge exchange collisions. At high energies these processes provide fast neutral beams that can be used for plasma heating. At low energies exchange collisions between ions and neutrals in a discharge result in energy losses by radiation following the decay of excited products. At this time there is a basic understanding of ion-atom charge exchange[1] but the same cannot be said for the more complex ion-molecule case. A model is proposed for ion-diatomic molecule charge exchange which is applicable in selected systems at low $\tau = E\theta$ (reduced scattering angle = beam energy x scattering angle). The model is a modification of one introduced to discuss collisions of the type $X^{++} + Y \rightarrow X^{+} + Y^{+}$ and is based on the large difference in the polarizabilities of the ground and excited states of atoms.[2] The model is applied to the $He^{+} + H_2$ system for which differential cross-section results on both the direct and exchange scattering are available.[3,4] This system is of current interest since it may play an important role in modeling planetary atmospheres.

Earlier work on $He^{+} + H_2$ at low keV energies has established several important features of the collision.[3,4] The following are of particular interest for this paper:

(1) Charge-exchange processes are important at small angles and dominate the scattering at larger angles. As an example at $E = 1.0$ keV and $\tau = 5$ keV deg. the probability of charge exchange is greater than 0.8.

(2) The behavior of the reduced cross-section as a function of reduced scattering angle strongly suggests that a common primary interaction may be responsible for both a strong peak in the direct inelastic scattering

and the small angle exchange scattering.[3] The direct channel (peak B in Ref. 3) corresponds to excitation of H_2 in a vertical transition to $H_2^*(a^3\Sigma_g^+,E,F,...)$ with a threshold inelastic energy loss $Q \cong 13$ eV. A time-of-flight measurement with modest energy resolution showed a peak in the He energy spectrum with a maximum at $Q \cong 13$ eV for small τ.[4]

The collision begins along the $He^+ + H_2(^1\Sigma_g^+)$ curve as shown in Fig. 1. At large interparticle separation the energy, to a good approximation, is given by

$$V = -\frac{\alpha e^2}{2r^4} \qquad (1)$$

where α is the polarizability of the H_2 target, e the charge of the incident He^+, and r the separation. The potential for this channel is plotted using $\alpha = 0.8 \times 10^{-24}$ cm^3 for H_2 [5] and includes only this term. A comparison of this curve with recent ab initio calculations[6] shows agreement to within a few tenths of an eV with those corresponding to He^+ parallel and perpendicular to the H_2 axis in the interparticle range in Fig. 1 and the corresponding figure (1) in Ref. 6. As may be seen in the figure, the $He^+ + H_2(^1\Sigma_g^+)$ curve crosses one corresponding to He $^3S(1s2s)$ + $H_2^{+*}(^2\Sigma_g^+$, v=10), associated with an energy loss Q = 13 eV reported for the small angle $He^+ + H_2 \rightarrow He + H_2^+$ collisions. This curve represents the system under a vertical transition from the initial H_2 to the final H_2^{+*} state and the crossing populates He ^3S. The potential for the excited state curve is obtained from eq. 1 using $\alpha = 46 \times 10^{-24}$ cm^3 and is displaced above the incident level by the excitation energy.[7] At large r the curve representing electron capture to He ^3S is almost degenerate with direct inelastic curves corresponding to $He^+ + H_2^*(a^3\Sigma_g^+,E,F,)$ and populates these direct channels at $Q \cong 13$ eV. The common primary interaction is the initial curve crossing. The above discussion assumed excitation of He ^3S. This is the least favorable case in Fig. 1 since it involves a triplet state which crosses at the smallest r. The results in Ref. 4 also allow for excitation of He $^1S(\alpha = 116 \times 10^{-24}\ cm^3)$.[7] This He(1s2s) curve is seen to cross the incident channel at larger r where the assumed potentials are more valid.

Figure 1 also shows curves corresponding to H_2^+ + He, vibrationally excited H_2^{+*} + He, and one for H_2^+ + He ^1S(1s2s). These curves are appropriately displaced from $He^+ + H_2$ and are plotted with $\alpha = 0.2 \times 10^{-24}\ cm^3$ for He($1s^2$).[7] Inspection of the figure shows that electron capture to He($1s^2$) will be weak compared to capture to He* since two crossings are required to reach the ground state. In H_2^+ + He collisions the crossing with the intermediate H_2^+ + He ^1S can populate $He^+ + H_2$ and can also result in collisional dissociation to H^+ + H + He at c.m. energies close to threshold.

At thermal energies (where a quantum mechanical treatment may be necessary) $He^+ + H_2$ can result in H^+ + H + He via the H_2^+ + He ^1S intermediate state, providing an additional mechanism for the low energy reaction. In $He^+ + D_2$ the dissociation cross-section is known to be smaller[8] and within this model can be attributed to the difference in centrifugal potential associated with the two collision systems. For a given impact parameter the distance of closest approach is smaller for H_2 than for D_2 allowing closer penetration into the critical region where the first crossing occurs.

Although short-range forces make significant contributions to the observed scattering in $He^+ + H_2$ all collisions must traverse the region in Fig. 1 at least once (highly excited exit channels lie above those shown). The curves in the figure represent selected states to illustrate the model. The proposed model uses simple potentials to indicate the importance of the long-range interaction in ion-molecule collisions. The excited channels in addition to being directly populated serve as intermediate states for other collision processes. The actual locations of crossings require detailed calculations of energy surfaces with particular attention to the long range forces. This simple model should be applicable to collisions involving other rare gas ions where the polarizabilities[9,10] of the excited states are large and to H^+ since the $n=2$ state of H has a linear Stark effect.

The author wishes to thank Professors R.B. Snyder and A. Russek as well as Dr. R.G. Garcia for useful discussions.

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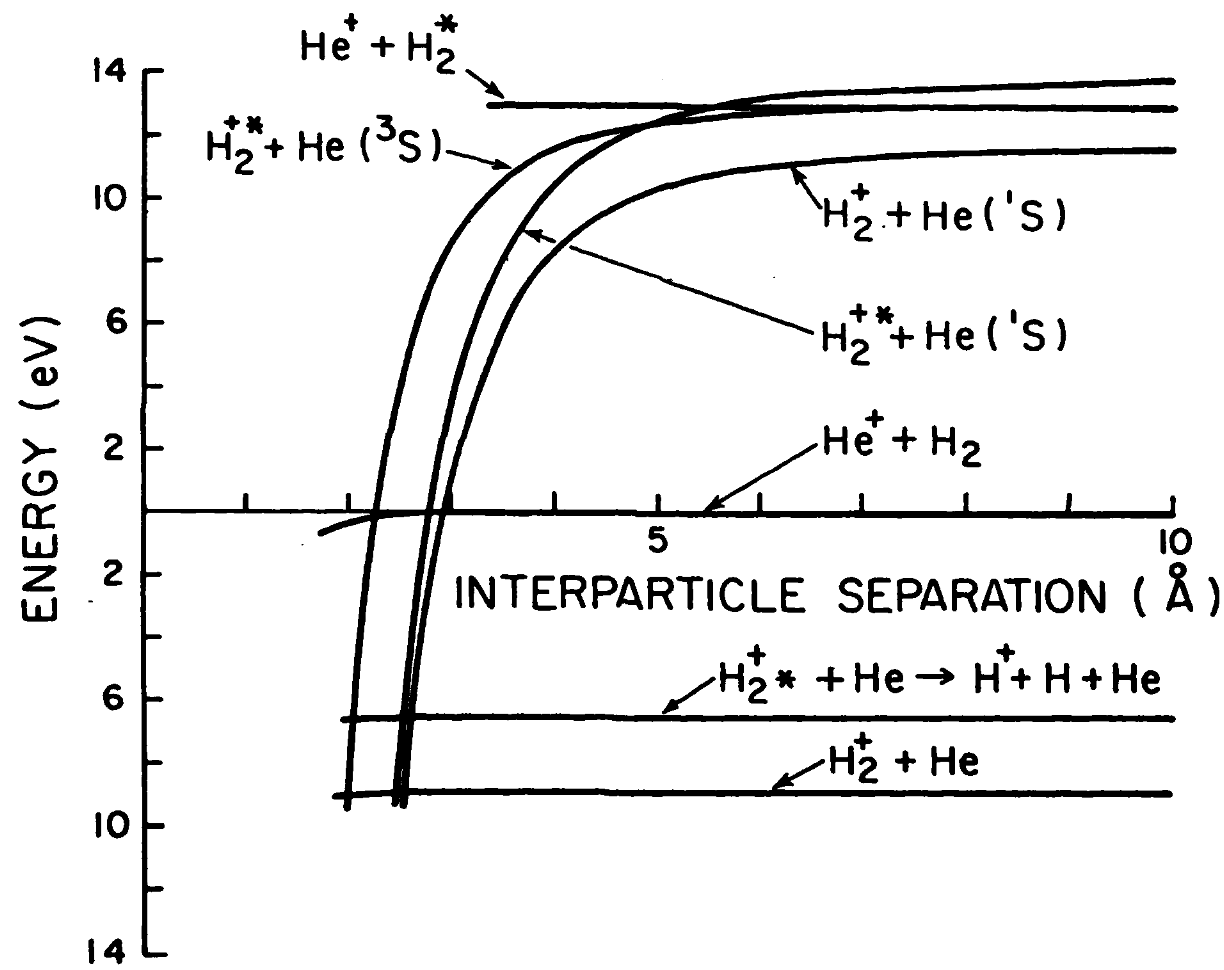
Fig. 1 Selected states in $He^+ + H_2$ and H_2^+ + He to illustrate the proposed model. Only states involving $H_2^+ \ {}^2\Sigma_g^+$ are shown since excitation of H_2^+ 2pσu should be weak in small τ collisions with $H_2 \ {}^1\Sigma_g^+$ molecules. The dominant charge exchange reported in references 3 and 4 is attributed to crossings between $He^+ + H_2({}^1\Sigma_g^+)$ and final states such as $H_2^{+*}({}^2\Sigma_g^+, v \approx 10)$ + He ${}^3S, {}^1S$ (1s2s). Couplings at large separation between these exchange states and $He^+ + H_2^*(a^3\Sigma_g^+, E, F, \ldots)$ result in direct inelastic scattering. $H_2^+ + He(1s^2)$ should only be weakly excited since two crossings are required. At thermal energies the intermediate $H_2^+({}^2\Sigma_g^+)$ + He ${}^1S$ (1s2s) state allows population of vibrationally excited $H_2^{+}* + He(1s^2) \rightarrow H^+ + H + He(1s^2)$ and $H_2^+ + He(1s^2)$. In $H_2^+ + He(1s^2)$ collisions couplings with the ${}^1S$ and ${}^3S$ states can result in collisional dissociation and charge transfer to $He^+ + H_2$ at energies near threshold.



$He^{+} + H_2^{*}$
$H_2^{+*} + He\ (^3S)$
$H_2^{+} + He\ (^1S)$
$H_2^{+*} + He\ (^1S)$
$He^{+} + H_2$
$H_2^{+*} + He \rightarrow H^{+} + H + He$
$H_2^{+} + He$
ENERGY (eV)
14
10
6
2
2
6
10
14
5
10
INTERPARTICLE SEPARATION (Å)

Abstracts, Int. Conf. on Atomic Physics, M.I.T. Press, August 1980.

Appendix III

HIGH RESOLUTION L X-RAY SPECTRA FROM MULTI-STRIPPED IONS IN 100 keV S^+ - Ar SINGLE COLLISIONS

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We report what we believe to be the first grazing-incidence spectral measurements of multiplet structure in the soft X-ray emission from keV-energy ion-atom single collisions (gas target). We discuss our observations of projectile X rays from S^+ - Ar collisions; preliminary spectra from the analogous P^+ - Ar system have already been reported.[1] The 100 keV high current (300 microamp/cm^2) mass-analyzed beam from the Univ. of Connecticut ion implantation facility was essential to the success of the experiment. Previous spectral measurements on P^+, S^+ and Ar^+ - Ar have been performed with curved-crystal Bragg spectrometers by the Livermore group.[2]

The objective was to obtain sufficient resolution in the soft X-ray spectrum under single-collision conditions to identify specific transitions from particular initial multiplets with multiple vacancies in the emitting ion. The coupling into multiplets of inner-shell vacancy states and outer-shell configurations following an ion-atom collision has a strong influence on the branching between X-ray and Auger electron emission.[3] The L X-ray fluorescence yield, ω_L, which is the ratio of decay by photon emission to all other decay modes[4], can vary for a low-Z element such as P, S or Ar from 10^{-4} to 1.0, depending on the initial post-collision L-shell hole configuration and the degree of excitation and ionization of the outer M-shell.[3] Within a given vacancy configuration of the excited S ion, Chen and Crasemann

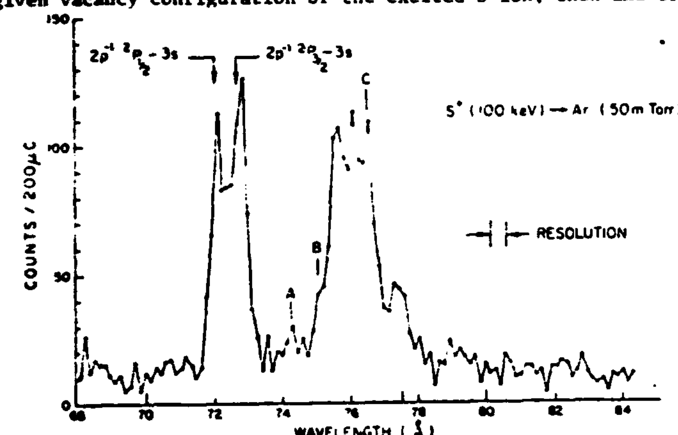

Figure 1. Portion of the grazing-incidence spectrum from 100 keV S^+ - Ar gas collisions. The 68-84 Å region shown has mostly neonlike and sodiumlike sulfur L-M X-ray transitions (range= 182-148 eV). The doublet identified arises from neonlike $S^{6+}[1s^2 2s^2 2p^5(^2P_{1/2},{}_{3/2})3s]$ fine-structure states, with ω_L=1 and wavelengths 72.029 and 72.663 Å. Transitions A, B, C, having fluorescence yields less than 1%, are discussed in the text.

calculate variations up to 2 orders of magnitude in ω_L depending on the angular-momentum coupling.[3] Non-statistical populations of $P_{3/2}$ and $P_{1/2}$ hole states have also been predicted and observed in certain heavy-particle collisions.[5] High resolution is necessary even for identification of the charge-state of the emitting projectile ions.

A new 1m Rowland circle grazing-incidence monochromator was used at 87° incidence on a 1200 groove/mm gold replica concave grating (first order diffraction efficiency at 44Å approx. 10%) with a gas-cell target chamber. Measurements in the 50-85Å region encompassed the Ar-L and S-L diagram lines and satellite spectral region. Low count rates dictated slit widths giving approx. 0.45Å FWHM resolution but the instrument is capable of nearly 3X higher resolution than this.

The 3s--$2p_{1/2,3/2}$ doublet identified near 72Å in Figure 1 is clearly separated. A "typical" lead stearate multilayer Bragg X-ray crystal we have tested had a resolution worse than 1.3Å FWHM and cannot resolve this doublet. The doublet and several other observed transitions have flourescence yields of unity (1.o). Transitions A (167 eV, from $2p^5 3s^2$), B(165 eV, from $2p^5 3s3p$) and C(162 eV, from $2p^5 3s^2 3p$) in Fig. 1 have calculated ω_L's less than .007, so that their excitation probability must exceed that of the 72Å doublet by at least 140X. All identified lines in both the P^+ -Ar and S^+ -Ar spectra are associated with high charge states: neonlike and sodiumlike projectile x-rays; there is little or no excitation of the higher Z Ar target, as expected. The phosphorus spectrum shows at least one transition identified as an "hypersatellite" with two 2p holes, consistent with ionization or excitation of both electrons in the 4fσ molecular orbital of the quasimolecule in the collision. Both P and S spectra show similar groups of lines of shorter wavelength than the 3s-2p doublet, involving excitation of the M shell: 4s--2p and 3d--2p lines.

Finally, we note that the statistical 2:1 ratio of the $P_{3/2}:P_{1/2}$ peaks is not observed in Fig. 1. Instead the ratio is approximately 1.3:1. Pure 4fσ excitation would lead to a final projectile state with m_l=0 and a 1:1 intensity ratio, so the data are not quite consistent with the simple single molecular-orbital mechanism for 2p hole excitation.

We acknowledge gratefully the support of the U.S. Army Research Office (Durham) together with supplementary support from the University of Connecticut Research Foundation. We thank Prof. J. Budnick for his efforts in acquiring the ion implantation accelerator.

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Appendix IV

MULTIPLET-RESOLVED L and L^2 SATELLITE X-RAY SPECTRA FROM 100 keV S^+ + Ar GAS COLLISIONS

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We recently reported high-resolution soft x-ray spectra from multi-stripped sulfur and phosphorus projectile ions excited in 80-150 keV collisions with low pressure Ar gas.[1] These single-collision spectra were taken in first order with a 1m grazing-incidence concave grating monochromator, using the high-current (~300uA/cm^2) mass-analyzed beam from the Univ. of Connecticut ion-implantation accele-

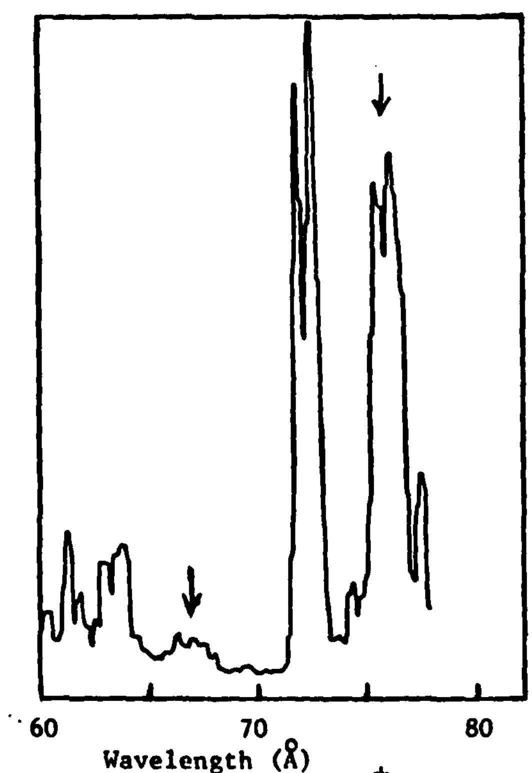

Fig. 1. Soft x-ray spectrum from 100keV S^+ + Ar gas collisions.

rator. A thin-window proportional counter was used as the detector.

The results, consistent with previous work by Fortner[2], permit for the first time the identification of many specific multiplets and charge states. Coupled with fluorescence yield calculations for specific multiple-vacancy multiplets[3], total cross sections for collisional excitation of a number of specific states can be determined. Figure 1 shows a typical L x-ray spectrum at approx. 0.45Å resolution. This is dominated by $L_{2,3,}$-M and $L^2_{2,3,}$-LM satellite lines corresponding to various high charge states up to fluorinelike S^{7+}, something of a surprise for 100 keV collisions. P^+ + Ar spectra at the same energy show analogous transitions.[1]

The S^{6+} $2p^5 3s$--$2p^6$ doublet at 72.03/72.66 Å has fluorescence yield $\omega_L = 1$, so there is no competing Auger transition. The group of lines between 74 and 80Å, representing initial configurations $2p^5 3s^m 3p^n$, (m + n = 2,3,4), have $\omega_L \lesssim 0.007$[3]; these are therefore much more strongly excited in the primary collision than the doublet. Little or no target excitation was found, consistent with the molecular-orbital picture for asymmetric collisions. There may be weak lines above 78Å up to the $L_{2,3}$-M diagram line.

The type of structure observed in both phosphorus and sulfur projectile spectra is very different from that seen with electron-excited spectra[4]. Preliminary wavelength measurements (see Fig. 1) for the following sulfur transitions, apparently not previously recorded, are as follows (identifications are tentative): $2p^4 3s^m 3p^n$ excitations ($m + n \geq 2$)-- 66.06, 66.66, 67.26, 67.78 and 68.32Å. Also, $2p^5 3s^m 3p^n$ excitations (m+n = 2,3)-- 74.46, 75.51, 76.26 and 77.46Å. Calibrations to ±0.07Å are with respect to known S^{q+} lines. Low fluorescence yields for all of the above imply strong excitation in collisions with Ar.

*Supported by the U.S.Army Res. Office and the U. of Ct. Res. Foundn.

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